# Experimental Limit to Interstellar $^{244}$Pu Abundance

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## ABSTRACT

Short-lived nuclides, now extinct in the solar system, are expected to be present in interstellar matter (ISM). Grains of ISM origin were recently discovered in the inner solar system and at Earth orbit and may accrete onto Earth after ablation in the atmosphere. A favorable matrix for detection of such extraterrestrial material is presented by deep-sea sediments with very low sedimentation rates (0.8–3 mm/kyr). We report here on the measurement of Pu isotopic abundances in a 1-kg deep-sea dry sediment collected in 1992 in the North Pacific. Our estimate of $(3 \pm 3) \times 10^5$ $^{244}$Pu atoms in the Pu-separated fraction of the sample shows no excess over the expected stratospheric nuclear fallout content and under reasonable assumptions sets a limit of 0.2 $^{244}$Pu atoms/cm$^2$yr for extra-terrestrial deposition. Using available data on ISM steady-state flux on Earth, we derive a limit of $2 \times 10^{-11}$ g-$^{244}$Pu/g-ISM for the abundance of $^{244}$Pu in ISM.


*Subject headings:* Nuclear reactions, nucleosynthesis, abundances– ISM: abundances–instrumentation: spectrographs–methods: laboratory

## 1. Introduction

The legacy of short-lived nuclides, now extinct in the solar system, is important since their production spans a smaller number of nucleosynthesis events than that of stable nuclides. Such events occur continuously in the Galaxy and it has been estimated (Meyer & Clayton 2000) that radioactive species with lifetimes $\gtrsim 5 - 10$ Myr should attain approximate steady-state abundances in the interstellar medium (ISM). The case of $^{244}$Pu ($t_{1/2}$=81 Myr) is interesting (Wasserburg et al. 1996), as a pure *r*-process nuclide produced in supernova and neutron-star disruption.

Its presence in early-solar material has been inferred from Xe isotopic anomalies (Rose and Kuroda 1965; Alexander et al. 1971; Wasserburg et al. 1985). The early-solar $^{244}Pu/^{238}U$ ratio was established to be ∼0.007 (Hudson et al. 1989) which is about the value expected from uniform production (Wasserburg et al. 1996).

The study of interstellar matter (ISM) has attracted considerable attention in the last decade, leading to the discovery of ISM grains in the inner solar system by the Ulysses (Grün et al. 1993) and Galileo space missions (Baguhl et al. 1995). These grains whose masses range from $10^{-15}$ to above $10^{-11}$ g are well identified as ISM by their retrograde trajectories, opposite to orbits of most interplanetary grains and close to that of interstellar gas (26 $kms^{-1}$ from respectively 253° and 5° ecliptic longitude and latitude), their constant flux in and out of the ecliptic plane and their high speeds in excess of the solar-system escape speed (Grün et al. 2000). Smaller-size particles are impeded from entering the planetary system by electromagnetic interactions with the ambient magnetic field. The elemental and isotopic composition of these grains will be a major goal of research with their recovery to Earth by the Stardust mission (Brownlee et al. 1996) planned for 2006. The modeling of the existing body of data from Galileo and Ulysses suggests that they can be matched only by assuming that there is no reduction of the interstellar dust flux at least as close as 1.3 AU and that up to 30% of the dust flux with masses above $10^{-13}$ g at these distances is of interstellar origin (Grün et al. 1997). More recently, even larger dust grains ($\sim 3 \times 10^{-7}$ g) incident on Earth and ablating in the atmosphere have been reliably identified as interstellar by their hyperbolic speeds and directions with the ground-based AMOR radar array (Baggaley 2000; Landgraf et al. 2000). An unmistakable signature for such material accreting on Earth would be the presence of short-lived nuclides now extinct in solar–system material.

We report on a search (Sakamoto et al. 2000; Valenta et al. 2000) for $^{244}Pu$

accumulated on Earth through cosmic dust deposition. This search is based on deep-sea sediment which, owing to very low sedimentation rates ($\sim$ 0.8–3 mm/kyr, see Goldberg and Koide 1962), offers a favorable matrix; on the other hand the nuclide $^{244}$Pu can be detected with high sensitivity by accelerator mass spectrometry (AMS) (Fifield 2000). Evidence for the presence of $^{244}$Pu in a rare-earth mineral was reported (Hoffman et al. 1971) but not confirmed since. The detection of short-lived nuclides injected from a hypothetical near-Earth supernova has also been considered (Ellis et al. 1996; Knie et al. 1999; Fields & Ellis 1999) and evidence for $^{60}$Fe in a deep-ocean ferromanganese crust, interpreted as originating from such an event, has been published (Knie et al. 1999). The same group recently reported measurements of Pu isotopes in a manganese crust (Wallner et al. 2000).

## 2. $^{244}$Pu analysis of a deep-sea sediment sample

A deep-sea sediment sample was treated at Kanazawa University for Pu extraction and $\alpha$ counting (Sakamoto et al. 2000). The sample (No. 92SAD01), originating from 80 kg of wet red clay, was dredged in 1992 from 5,800 m in water depth in the Pacific Ocean (9°30'N, 174°18'W) over a layer of seabed of $\sim$ 2 m$^2$ area and $\lesssim$ 0.3 m sediment depth. After washing and filtering off nodules, an aliquot of 1.054 kg of the resulting 12.85 kg dry sediment was used. $^{239,240}$Pu of nuclear-bomb fallout origin present in the sample was used throughout as an in-situ tracer for the $\alpha$ and AMS analysis. Pu was extracted by alkali fusion and anion exchange and electrodeposited on a stainless steel disk. A 34 g aliquot of the dry-sediment sample was processed separately after addition of a $^{242}$Pu tracer to determine the chemical efficiency of Pu extraction ($\sim$40%). A value of 34$\pm$3 $\mu$Bq-$^{239+240}$Pu/g was determined for the dry sediment from $\alpha$ counting of the latter sample and two independent $\alpha$ measurements. A 546-day $\alpha$-counting of the main Pu fraction was

performed using an $\alpha$ spectrometer with an efficiency of ∼25%. The dominant $\alpha$ group observed corresponds to $^{239+240}$Pu with an activity of ∼0.013 Bq or ∼$1.1\times10^{10}$ $^{239+240}$Pu atoms.

After $\alpha$-counting, the Pu-electrodeposited disk of the main fraction was shipped to the Hebrew University for AMS analysis. The Pu layer was dissolved in a HCl solution containing 2.4 mg $Fe^{3+}$ and Pu co-precipitated with iron (III) hydroxide using ammonia solution. The overall efficiency of Pu recovery was determined to be ∼80%. The precipitate was dried, ignited to $Fe_2O_3$, divided in two parts, then pressed into the cathode holders of a Cs-sputter negative ion source (Gelbart et al. 1997). For normalization of the AMS analysis, calibration cathodes containing $(3.2\pm0.3)\times10^{10}$ $^{244}$Pu atoms were prepared with the same procedure after spiking with a $^{244}$Pu solution (0.32 ng-$^{244}$Pu/mL) prepared at Argonne National Laboratory. The AMS analysis was performed at the Koffler 14UD Pelletron tandem accelerator (Weizmann Institute) with the setup described in detail in Berkovits et al. 2000. $^{A}$Pu (A = 239,240,241,242,244) were sequentially analyzed by injecting $^{A}PuO^{-}$ into the accelerator. After acceleration with a terminal voltage of 7.1 MV, $^{A}Pu^{9+}$ ions were momentum and velocity-analyzed before entering a detection system determining time of flight and energy. Careful calibration of the voltages of the accelerator and of the Wien filter were required to analyze the Pu isotopes through the beam transport system, keeping magnetic rigidity constant. Unambiguous identification of $^{A}$Pu ions was obtained in the detector from the time-of-flight and energy signals.

Fig. 1a shows the Pu isotopic distribution measured by AMS for the calibration cathodes, in good agreement with a distribution determined by $\alpha$ spectrometry. The cathodes were run to exhaustion of the Pu output (∼ 4 hours), yielding an overall detection efficiency (counts/atoms in cathode) of ∼$3\times10^{-6}$. The measured isotopic distribution for the sediment sample is shown in fig. 1b. The average count rate for $^{239}$Pu over the lifetime

of the cathode was 2.2 counts per second. The absolute number of combined $^{239+240}$Pu atoms, determined in the AMS measurement using the above efficiency, is $1.4\times10^{10}$, in reasonable agreement with the $\alpha$-counting value. The measured ratio $^{240}$Pu/$^{239}$Pu is 0.15±0.02, in agreement with the stratospheric fallout value 0.18±0.01 (see Cooper et al 2000). Reasonable agreement is also observed for $^{241}$Pu after correction for its radioactive decay. The measured relative abundance of $^{242}$Pu is in excess of the expected value for fallout or any natural source and is attributed to a possible contamination originating in the use of a $^{242}$Pu tracer in the laboratory. The high isotopic purity of the tracer ($> 90\%$) precludes any significant effect on other Pu isotopes. Special care was taken to reduce any memory effect in the AMS ion source between the measurement with a calibration cathode and that of the sediment sample. A run of 4 hours before the latter measurement, using a stainless steel dummy cathode in the ion source, yielded no counts of $^{244}$Pu. Only one count of $^{244}$Pu with no background ions was detected for the sediment sample during a 3.5-hour measurement, corresponding to $(3 \pm 3) \times 10^5$ $^{244}$Pu atoms in the cathode. The $^{244}$Pu abundance in stratospheric nuclear fallout is not known and we estimate it here by extrapolating measured $^{A+1}$Pu/$^{A}$Pu (A = 239–241) fallout ratios (see Cooper et al. 2000). We use in this extrapolation the experimental distribution of Pu isotopes (A = 239–244) determined in the Mike thermonuclear explosion (Diamond et al. 1960) and assume that the effective neutron capture cross sections in this experiment are the same as for the global nuclear atmospheric tests. The double ratio $(^{244}\mathrm{Pu}/^{242}\mathrm{Pu})_s/(^{244}\mathrm{Pu}/^{242}\mathrm{Pu})_M$ is then given by $(<\Phi>_s/<\Phi>_M)^2$, where $s$ and $M$ denote respectively stratospheric and Mike tests and $<\Phi>$ denotes the effective neutron fluence. The ratio $<\Phi>_s/<\Phi>_M$ obtained from the data for $^{A+1}$Pu/$^{A}$Pu (A = 239–241) is $0.52 \pm 0.12$. The fallout–$^{244}$Pu content of our sample is thus estimated as $(7 \pm 3) \times 10^5$ atoms. We conclude from a comparison with our measurement that any excess $^{244}$Pu accumulated in deep-sea sediment from extraterrestrial sources is $< 1 \times 10^6$ $^{244}$Pu atoms/kg–dry sediment at a 90% confidence level.

## 3. Limits on $^{244}$Pu abundance

The fact that no $^{244}$Pu signal is observed above the nuclear-fallout value is significant in that it helps to place a bound on the $^{244}$Pu abundance in the ISM. We shall take $r_{sed} =$ 1 mm/kyr as a representative value for the sedimentation rate in deep-sea pelagic sediment (Goldberg & Koide 1962). Based on the short residence times of Pu in the atmosphere and in the ocean ($< 30$ yrs, Lal 2001), we can assume that the steady-state accumulation rate of ISM–$^{244}$Pu in deep-sea sediment is equal to its accretion rate on Earth. Using the above limit for $^{244}$Pu concentration $C_{sed}^{244}$ in the sediment, a sediment density $\rho_{sed} =$ 1.5 g/cm$^3$, we obtain a limit for the steady-state interstellar $^{244}$Pu deposition on Earth $\phi_{\oplus}^{244} = 10^{-7} C_{sed}^{244} \rho_{sed} r_{sed} < 0.2$ $^{244}$Pu atoms/cm$^2$ yr.

The ISM accretion flux onto Earth is highly uncertain at present. A quantitative estimate may however be attempted using the recent experimental AMOR data (Baggaley 2000; Landgraf et al. 2000) which determine a flux $\phi_{1AU}^{ISM}$ of $\sim 1.8 \times 10^{-12}$ and of $< 3 \times 10^{-14}$ cm$^{-2}$s$^{-1}$ respectively at southern and northern ecliptic latitudes, for ISM grains of mass $m_{grain}^{ISM} > 3 \times 10^{-7}$ g. These values roughly fit, with a $m^{-1}$ power law, the mass distribution of the interstellar grains identified by the Ulysses mission (Landgraf et al. 2000). The flux $\phi_{1AU}^{ISM}$ is highly anisotropic and, assuming that unidirectional particles ablating on the Earth's cross section average homogeneously over the Earth's surface, we obtain an ISM steady-state deposition flux $\phi_{\oplus}^{ISM} = \phi_{1AU}^{ISM} m_{grain}^{ISM}/4 > 1.4 \times 10^{-19}$ g-ISM/cm$^2$s. The corresponding limit on the abundance of $^{244}$Pu in ISM is

$$C_{ISM}^{244} = 3.2 \times 10^{-8} (\phi_{\oplus}^{244}\ 244/\mathcal{N}_{\mathcal{A}})/\phi_{\oplus}^{ISM} < 2 \times 10^{-11}\ \text{g-}^{244}\text{Pu/g-ISM}. \qquad (1)$$

Further assuming that ISM has an early solar-system uranium abundance of $1.6 \times 10^{-8}$ g-U/g (Anders & Grevesse 1989), we derive a limit of $(^{244}Pu/U)_{ISM} < 1 \times 10^{-3}$ in steady-state ISM. This upper limit is smaller by about one order of magnitude than the adopted early-solar ratio $(^{244}Pu/U)_{\odot} = 0.007$. The latter value was shown to be about half of that

calculated with a uniform production (UP) model (Wasserburg et al. 1996) of actinides by supernova $r$-process and it may be significant that the $(^{244}Pu/U)_{ISM}$ limit estimated here lies even lower. Wallner et al. 2000 recently reported on a $^{244}$Pu measurement in a 120 g ferromanganese crust where they detected one count of $^{244}$Pu corresponding to $\sim 1 \times 10^5$ $^{244}$Pu atoms. The authors derive from a known accumulation age of the crust ($\sim$ 13 Myr), a flux of $\sim$ 300 $^{244}$Pu atoms/cm$^2$Myr and conclude that the ratio between this flux and the flux of $^{60}$Fe measured in a similar deep-ocean crust is compatible with the signal of a near-Earth ($\sim$ 30 pc) supernova explosion $\sim$ 5 Myr ago (Knie et al. 1999). It is interesting to interpret this $^{244}$Pu flux estimate in terms of steady-state ISM deposition rather than effects of ejecta from a near-Earth supernova. Using the assumptions and estimates detailed above for the ISM steady-state accretion flux onto Earth, the flux determined by Wallner et al. would lead to a ratio $(^{244}Pu/U)_{ISM} \lesssim 2 \times 10^{-6}$, smaller by three orders of magnitude than the early-solar value or the UP model. We do not know at this point whether the low $(^{244}\mathrm{Pu/U})_{ISM}$ values estimated in the present work reflect (*i*) an overestimate of the steady-state ISM flux onto Earth, (*ii*) a possible fractionation process reducing the Pu transport to Earth or, for the ferromanganese crust, reducing the Pu intake or (*iii*) perhaps, represent a genuine low $^{244}$Pu abundance in ISM.

The experimental limits derived in the present work were constrained by the nuclear-fallout Pu content of the deep-sea sediment sample. This ensured that Pu geochemical and chemical behaviors were under control; however, one could establish more stringent limits by using a deep-sea core sample that is clean of artificial Pu input and by improving the overall sensitivity of the $^{244}$Pu detection. We are presently pursuing these directions.

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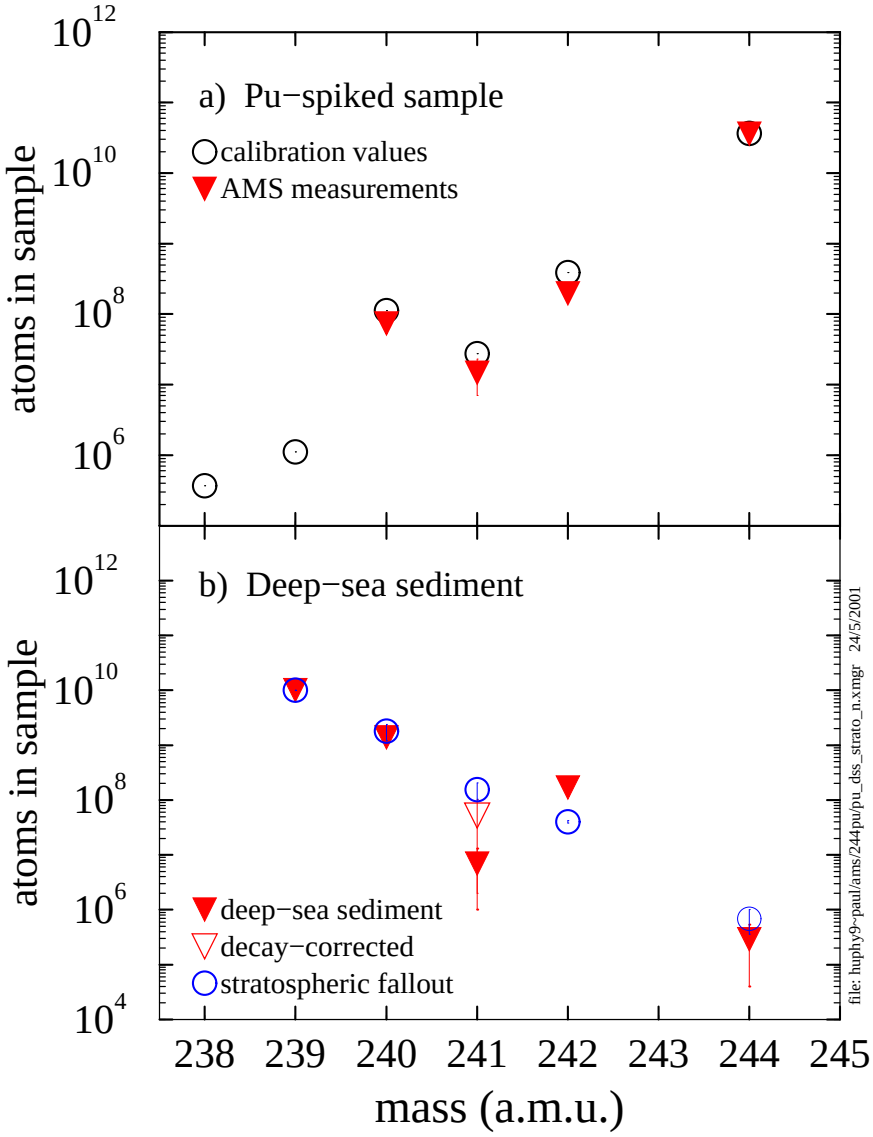


Fig. 1.— Isotopic distribution of plutonium (solid triangles) measured by accelerator mass spectrometry (AMS) for : *a)* a calibration sample containing $3.7 \times 10^{10}$ $^{244}$Pu atoms. The open circles show the distribution measured by $\alpha$-counting of a thin source prepared from the same solution as the AMS calibration sample. The AMS results were normalized to the number of $^{244}$Pu atoms obtained from the $\alpha$ counting. *b)* the plutonium fraction extracted from the 1.020-kg deep-sea dry sediment 92SDA01. The AMS results were normalized to the combined number of $^{239,240}$Pu atoms measured by $\alpha$ spectrometry. The open circles represent the expected Pu isotopic distribution of nuclear-bomb stratospheric fallout. The open triangle for A=241 includes the radioactive decay correction of fallout $^{241}$Pu. See text for details.